Temperature-induced metal-insulator transition in a narrow-band model with non-equivalent Hubbard subbands at half-filling

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Abstract

In the present paper the temperature-induced metal-insulator transition is studied in a generalized Hubbard model with correlated hopping using recently obtained expression for energy gap. The dependence of energy gap on concentration of doubly occupancy leads to increasing energy gap width with increase of temperature. Thus narrow-band system can undergo transition from a metallic state to an insulating state with the increase of temperature.

For some values of intra-atomic Coulomb repulsion U and w_0 (w_0 is half-bandwidth without taking into account of correlated hopping of electrons) we find the values of temperature when narrow-band material undergoes transition from a metallic state to an insulating state. We show that at given U/w_0 metal-insulator transition in model with non-equivalent Hubbard sub-bands can occur at smaller temperature than in the Hubbard model. It testifies on the fact that taking into account of correlated hopping is important for a consideration of metal-insulator transition problem.

The obtained results are applied to the interpretation of the experimental data.

1 Introduction

It is known that the electron-hole symmetry is peculiar to the Hubbard model [1]. One of manifestations of this symmetry is the equivalence of the lower and upper Hubbard bands. It is the result of an equality of hopping integrals describing both "translational" hoping of holes and doubly occupied sites (doublons) and the processes of their paired creation and destruction. Equality of noted hopping integrals is caused by neglecting the matrix elements of electron-electron interaction

in the general Hamiltonian (the matrix elements (1) describe hopping of electrons between i and j lattice sites; ϕ-function is the Wannier function).

However, theoretical analysis, on the one hand, and available experimental data, on the other hand, point out the fact that the Hubbard model generalization by taking into account correlated hopping (1) is of principle necessary [2] – [5]. In such model hopping integrals describing "translational" hopping of holes and doublons are different. These hopping integrals also differ from the hopping integral which is connected with the processes of paired creation and destruction of holes and doublons. In consequence of that the lower and upper Hubbard bands are non-equivalent (non-symmetric). In recent years similar models have been studied intensively [6] – [11].

Important puzzle arising in an investigation of these models is metal-insulator transition problem which is one of the most essential in narrow-band physics [12] – [14]. In this connection special interest is challenged by the observable metal-to-insulator transitions in some narrow-band materials with the increase of temperature (see, for example [14] – [18]).

On the basis of an approach proposed in the papers [3, 19] we have studied metal-insulator transition in a model of narrow-band material with non-equivalent Hubbard subbands (so-called "non-symmetric Hubbard model") at half-filling and zero temperature in the paper [20]. The present paper is devoted to a further study of metal-insulator transition in a model with non-equivalent Hubbard subbands, in particular, an investigation of temperature-induced metal-to-insulator transition.

2 Results

We start from the following natural generalization of the Hubbard model [1] at half-filling including correlated hopping (1) [2, 3, 20]

$$\begin{aligned} H = \quad & -\mu \sum_{i\sigma} a_{i\sigma}^{+} a_{i\sigma} + (t_0 + T_1) {\sum_{ij\sigma}}' a_{i\sigma}^{+} a_{j\sigma} + T_2 {\sum_{ij\sigma}}' \left(a_{i\sigma}^{+} a_{j\sigma} n_{i\bar{\sigma}} + h.c. \right) \\ & + U \sum_{i} n_{i\uparrow} n_{i\downarrow}, \end{aligned} \tag{2}$$

where μ is the chemical potential, $a_{i\sigma}^{+}, (a_{i\sigma})$ is the creation (destruction) operator of an electron of spin σ ($\sigma = \uparrow, \downarrow$) on i-site ($\bar{\sigma}$ denotes spin projection which is opposite to σ), $n_{i\sigma} = a_{i\sigma}^{+} a_{i\sigma}$ is the number operator of electrons of spin σ on i-site, U is the intra-atomic Coulomb repulsion, $t_0,\ T_1,\ T_2$ are the integrals of electron hopping between nearest neighbors,

$$T_1 = \sum_{\substack{k \neq i \\ k \neq j}} J(ikjk), \quad T_2 = J(iiij);$$

the primes at the sums in Hamiltonian (2) signify that $i \neq j$.

Using a generalized mean-field approximation [3, 19] in Green function method we obtain for a paramagnetic state the single-particle energy spectrum as [20]

$$E_{1,2}(\mathbf{k}) = -\mu + \frac{(1-2d)(t_{\mathbf{k}} + \tilde{t}_{\mathbf{k}}) + U}{2} \mp \frac{1}{2} F_{\mathbf{k}}, \tag{3}$$

where $\mathbf{k}$ is the wave vector, $E_1(\mathbf{k})$, $(E_2(\mathbf{k}))$ are the energies of electrons within the lower (upper) Hubbard band, d is the concentration of polar states (holes or doublons), $t_{\mathbf{k}}$, $\tilde{t}_{\mathbf{k}}$, $t'_{\mathbf{k}}$ are the Fourier transforms of respective hopping integrals $t = t_0 + T_1$, $\tilde{t} = t + 2T_2$, $t' = t + T_2$; t and $\tilde{t}$ are terms describing hopping of quasiparticles within the lower and upper Hubbard bands (hopping of holes and doublons) respectively, t' describes quasiparticle hopping between hole and doublon bands (the processes of paired creation and destruction of holes and doublons).

Note that single-particle energy spectrum (3) gives the exact atomic and band limits: if $U = 0$ and $t_{\mathbf{k}} = \tilde{t}_{\mathbf{k}} = t'_{\mathbf{k}} = t_0(\mathbf{k})$ (neglecting correlated hopping) then $E_{1,2}(\mathbf{k})$ take the band form, if $t_{\mathbf{k}} = \tilde{t}_{\mathbf{k}} = t'_{\mathbf{k}} \to 0$ then we obtain exact atomic limit.

With the help of single-particle energy spectrum (3) we find the expression to calculate the energy gap width (difference of energies between bottom of the upper and top of the lower Hubbard bands):

$$\Delta E = -(1-2d)(w+\tilde{w}) + \frac{1}{2}(Q_1 + Q_2), \tag{5}$$

$$Q_1 = \sqrt{\left[B(w-\tilde{w}) - U\right]^2 + (4dzt')^2}, \tag{6}$$

$$Q_2 = \sqrt{\left[B(w-\tilde{w}) + U\right]^2 + (4dzt')^2}, \tag{7}$$

where w and $\tilde{w}$ are halfwidths of the lower (hole) and upper (doublon) Hubbard bands: $w = z|t|$, $\tilde{w} = z|\tilde{t}|$ (z is the number of nearest neighbors to a site).

At given w, $\tilde{w}$, t', U and change of d (which can be caused by external influences, in particular, by temperature) energy gap (5) vanishes when the condition $d \le d_0$ is satisfied where d_0 is the root of the equation

$$-(1-2d)(w+\tilde{w}) + \frac{1}{2}(Q_1 + Q_2) = 0. \tag{8}$$

Then a metallic state ($\Delta E \le 0$) is realized at $d \le d_0$, an insulating state ($\Delta E > 0$) is realized at $d > d_0$.

If $t = \tilde{t} = t' = t_0$ (the Hubbard model) then energy gap (5) takes the following form [19]:

$$\Delta E = -2w(1-2d) + \sqrt{U^2 + (4dw)^2}, \tag{9}$$

and vanishes when the condition $d \le d_0$ is satisfied where

$$d_0 = \frac{1 - (U/2w)^2}{4} \qquad (2w \ge U). \tag{10}$$

From Eq. (8) we obtain the dependence of d_0 on U/w ratio (Fig. 1). The parameters $\tau_1 = T_1/|t_0|$, $\tau_2 = T_2/|t_0|$ characterize value of correlated hopping. From Fig. 1 one can see that value of d_0 depends on the parameters of correlated hopping τ_1, τ_2 (thus on $\tilde{w}/w$) weakly when U/w is near zero. But with the increase of U/w the value of d_0 begins to depend strongly on the parameters τ_1, τ_2 . Fig. 1 points out the extension of region in which narrow-band material is a metal with the increase of $\tilde{w}/w$ ratio (at given U/w).

The concentration of polar states (obtained with the help of the Green function $\langle\langle a_{i\sigma} n_{i\bar{\sigma}} | a^+_{j\sigma} \rangle\rangle$) is [20]

$$\frac{1}{N}\int_0^w \left[\frac{C}{\quad} + \frac{D}{\quad} \right]$$

with

$$C_\varepsilon = \frac{1}{2} - \frac{U}{2F_\varepsilon} - \frac{B\varepsilon}{2F_\epsilon}\left(\frac{\tilde{t}}{t} - 1\right), \tag{12}$$

$$D_\varepsilon = \frac{1}{2} + \frac{U}{2F_\varepsilon} + \frac{B\varepsilon}{2F_\varepsilon}\left(\frac{\tilde{t}}{t} - 1\right), \tag{13}$$

and chemical potential of narrow-band model with non-equivalent Hubbard subbands is given by the equation

$$\int_{-w}^{w}\left[\frac{1}{\exp\frac{-E_2(\varepsilon)}{k_BT}+1} - \frac{1}{\exp\frac{E_1(\varepsilon)}{k_BT}+1}\right]d\varepsilon = 0, \tag{14}$$

$E_{1,2}(\varepsilon),\ F_\varepsilon$ are obtained from respective formulae (3), (4) for $E_{1,2}(\mathbf{k}),\ F_\mathbf{k}$ by substitution of $t_\mathbf{k} \to \varepsilon,\ \tilde{t}_\mathbf{k} \to (\tilde{t}/t)\varepsilon,\ t'_\mathbf{k} \to (t'/t)\varepsilon$. Here we have used the rectangular density of states

$$\frac{1}{N}\sum_\mathbf{k}\delta(E - t(\mathbf{k})) = \frac{1}{2w}\theta(w^2 - E^2), \tag{15}$$

where $\theta(x) = 1$ if $x > 0,\ = 0$ otherwise; N is the number of lattice sites.

3 Discussion

The temperature dependence of chemical potential of a narrow-band model with non-equivalent Hubbard subbands obtained from Eq. (12) is plotted in Fig. 2. One can see that in the considered model in the region of low and normal temperatures chemical potential is essentially dependent not only on the parameters w and $\tilde{w}$ and also on temperature (in contrast to the Hubbard model where $\mu = U/2$), and what is more with the decrease of temperature chemical potential rapidly increases depending on the paramaters of non-equivalence of Hubbard bands $\tau_1,\ \tau_2$. In high temperature region in the proposed model chemical potential tends to $U/2$ with the increase of temperature; really, at $T \to \infty$ the probabilities of an electron finding within the lower and upper Hubbard bands (independently of their bandwidths ratio) are equal.

At given $U,\ w,\ \tilde{w},\ t'$ (constant exterior pressure) concentration of polar states (11) increases with the increase of temperature. It leads to the fact that system can undergo transition from the state with $\Delta E \leq 0$ to the state with $\Delta E > 0$, i.e. metal-to-insulator transition can occur. In this case the results obtained in the Hubbard model and those obtained in non-symmetric Hubbard model can be essentially different (Fig. 3 illustrates it). Let us take for example $U/w = 0.9$. One can see that at $T = 0$ the energy gap width in both models is $\Delta E < 0$ (a metallic state). With the increase of temperature metal-to-insulator transition does not occur in the Hubbard model, in non-symmetric model the values of parameters $\tau_1,\ \tau_2$ exist at which metal-to-insulator transition occurs.

In case metal-to-insulator transition occurs in both models from Fig. 3 one can

model. So, for example, for $w_0 = z|t_0| \approx 1.05$ eV (such bandwidth of NiS_2 was estimated in paper [16]) in considered model in a paramagnetic state metal-to-insulator transition occurs at $T \approx 280$ K for $U/w_0 = 1.94$ and $\tau_1 = \tau_2 = 0.01$ (observable the transition temperature of NiS_2 is $T \sim 280$ K at $p \sim 3$ MPa [17]). For the same value of U/w_0 metal-to-insulator transition occurs at $T \approx 940$ K when $\tau_1 = \tau_2 = 0$ (neglecting of correlated hopping, the Hubbard model) and at $T = 0$ K when $\tau_1 = \tau_2 = 0.015$. If $U/w_0 = 1.98$ then transition from a metallic state to an insulating state is realized at $T \approx 290$ K for $\tau_1 = \tau_2 = 0$; at $T = 0$ K when $\tau_1 = \tau_2 = 0.005$. Note that at $U \sim 2w$ the temperatures of metal-to-insulator transiton found in both models are essentially different; with a deviation from this ratio the difference decreases.

The obtained temperature dependence of energy gap can explain observable transition from the state of a paramagnetic metal to the paramagnetic Mott-Hubbard insulator state in the $(V_{1-x}Cr_x)_2O_3$ compound [14], [15] in NiS_2 [17] and in the $NiS_{2-x}Se_x$ system [17, 18] with the increase of temperature.

In summary, metal-to-insulator transition observable in the materials with narrow energy bands can be explained on the basis of the proposed approach at realistic values of the parameters characterizing non-equivalence of the Hubbard subbands. In this way series of results obtained from such consideration are essentially distinct from those obtained without taking into account this non-equivalence (the Hubbard model). In particular, the temperature of metal-to-insulator transition is essentially smaller than in the Hubbard model (this fact agrees to the observable transition temperature), and chemical potential disctinguishes from the value $\mu = U/2$ (obtained in the Hubbard model) and is temperature dependent.

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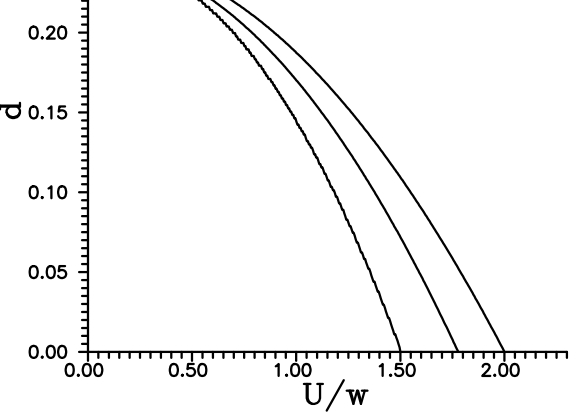
Figure 1: The dependence of d_0 on U/w ratio: the upper curve corresponds to $\tau_1 = \tau_2 = 0$ (the Hubbard model); the middle curve – $\tau_1 = \tau_2 = 0.1$; the lower curve – $\tau_1 = \tau_2 = 0.2$. To the left of respective curve there is a metallic phase, to the right – an insulating phase.

Figure 2: The temperature dependence ($\theta = k_B T$) of chemical potential μ for $U/2w = 1$: the upper curve corresponds to $\tau_1 = \tau_2 = 0.3$; the lower curve – $\tau_1 = \tau_2 = 0.1$; the straight line corresponds to values of chemical potential in the Hubbard model ($\tau_1 = \tau_2 = 0$).

Figure 3: The dependence of energy gap on temperature at $U/w = 0.9$. The upper curve corresponds to $\tau_1 = \tau_2 = 0.2$, the middle curve – $\tau_1 = \tau_2 = 0.1$, the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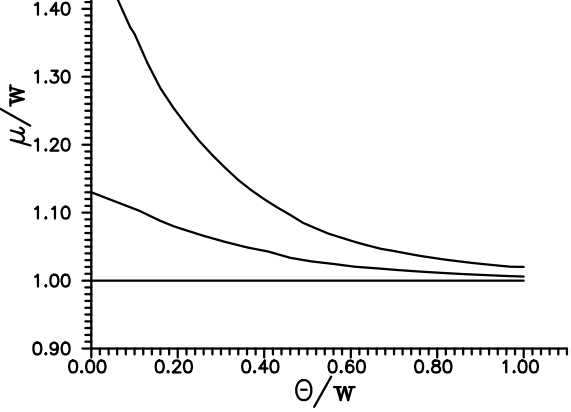
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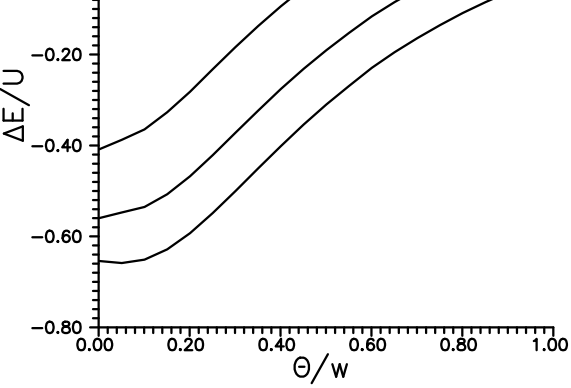
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